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PATENT SPECIFICATION

DRAWINGS ATTACHED

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COMPLETE SPECIFICATION

Improvements in or relating to the Bonding of a Ceramic Part to a Metallic Part

We, CERBERUS A.G., a Swiss Company, of Mannedorf, Switzerland, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to the bonding of a ceramic part to a metallic part.

The mechanically firm and hermetic bonding of metallic bodies to ceramic materials assumes ever greater technical importance. Owing to their good electrical properties in the high-frequency field and their superior heat resistance, interest has of late largely centered on oxide ceramic materials, such as aluminium oxide or beryllium oxide. The thermal expansion of these oxide ceramics is determinable, and it has been the practice to find a metal associate the coefficient of expansion of which largely corresponds to that of the ceramic material. This has been extremely important as all oxide ceramic materials are inelastic, that is to say, possess only low bending and tensile strength combined with great hardness and compressive strength. In addition, the soldering temperatures are advantageously 900° C and higher so that even small differences in the coefficients of expansion will cause conditions of heavy stress due to the great difference between room and soldering temperatures.

Practice has shown that it is very difficult to find pure metals or one of the special vacuum-type alloys which come sufficiently close to the desired properties. The so-called fused alloys, particularly those forming the iron-nickel-cobalt group, are quite adequate up to temperatures of 400 or 600° C. The break in the expansion characteristic appearing in all ferro-magnetic materials is located at this temperature level so that application at higher temperatures still becomes fairly problematic. These difficulties are commonly avoided by providing resilient or spring action for the metal parts to be soldered by particular configuration thereof or by employing very thin units. On the other hand, the expansion characteristic of pure oxide ceramics may be influenced or controlled in a desired manner by application of additives. Although the coefficients of expansion may, thus, be largely adjusted between metal and ceramic material, the quality of the ceramic material is most frequently adversely affected.

According to the present state of the art, there are two basic methods of obtaining mechanically firm and hermetic bonding. In one of these entailing application of a so-called two-phase method, the ceramic material is initially coated with a very thin metal film by applying a suspension of metal or metal hydride powder to the ceramic material, and is then caused to react under certain conditions after drying. Whether the metal powder reacts directly with the oxide ceramic body or only with a binder still present therein, is of no particular moment in this connection. The thus obtained firmly bonded film is commonly thickened by galvanising. A third step comprises the soldering of the premetallised ceramic material to the metal. Since the working temperatures of the commercially available solders form a practically continuous series from 150° C to 1500° C, it is thus readily possible to obtain any soldering temperature in order to reduce thermal stresses as far as possible. Moreover, selection of a ductile solder enables residual stresses between the ceramic material and the metal to be largely taken up within the region of the soldering zone. This last mentioned feature constitutes the essential advantage of the hereinabove described method.

[Price 4s. 6d.]

In the other so-called single-phase methods, the ceramic material is metallised and soldered to the metal body in a single operation. Only such metal or metallic components may be used for metallising the ceramic material which are able to react directly with the latter. These are substantially the high-melting metals of Group IVa through VIIa of the Periodic Table of Elements, which are subsequently briefly referred to as "reactive" metals.

Soldering must be effected at temperatures lower than the fusing temperatures of the components to be bonded. This is the reason why the reactive metal must be obtained in a highly reactive form. This is achieved, by way of example, by dissolving or suspending it in a liquid metal associate or partner so that it may, for example form a eutectic. The quantity of the reactive metal in the soldering combination may not be below a certain minimum in order to enable bonding to the ceramic material to be obtained. This minimum quantity would in general be sufficient to cause hardening and embrittlement of the entire solder so that the stresses arising from differences between the coefficients of expansion of the ceramic material and the metal would possibly cause destruction of the bond.

The present invention avoids both the difficulties caused by defective adjustment of the expansion characteristics and the complicated steps of the two-phase method. Bonding is performed in a single operation and no adjustment of the expansion characteristics of the metal and the ceramic material is necessary.

According to the present invention, there is provided a method of bonding a ceramic part to a metallic part, comprising disposing between said ceramic part and said metallic part first and second different metallic layers such that the first layer, which comprises a first metallic material capable of reacting with, and bonding itself to, said ceramic part, is disposed adjacent said ceramic part and that the second layer, which comprises a second metallic material capable of bonding itself to said metallic part, is disposed adjacent to said metallic part, heating the ceramic part, the metallic part and the layers in an inert atmosphere or in vacuo so as to bond the ceramic part to the metallic part, and causing there to be present during the heating operation a solid metallic separating layer separating molten, first metallic material adjacent to said ceramic part from molten, second metallic material adjacent to said metallic part, eutectics being formed between said separating layer and said molten, first metallic material and between said separating layer and said molten, second metallic material or said metallic part, the heating being performed at a temperature lower than the melting point of said separation layer and higher than the melting point of each of said eutectics.

It is possible to have between the first and second metallic layers at least one further metallic layer.

possible 3rd intermediate layer

The bond formed between the ceramic part and the metallic part is harder in the zone adjacent to the ceramic part than in the zone adjacent to the metallic part, since distribution in the latter zone of the reactive metal causing hardening and embrittlement is prevented by the separating layer.

intermediate layer is optional

The heating is performed in an inert atmosphere, for example an inert gas such as argon, or *in vacuo* in order to avoid oxidation or nitriding, for example, of the various metallic substances present.

This heating can be performed, by way of example, in a suitable furnace provided with electrical filament winding or by high-frequency heating, with the use of suitable measuring instruments enabling the furnace or the like to be controlled.

Ceramic materials particularly suitable for the purpose are highly pure sintered ceramic types such as aluminium or beryllium oxide, which are commercially available in degrees of purity of 97% and higher. Among the aforesaid group of reactive metals, titanium and zirconium are particularly advantageous for economic and practical considerations. When a ceramic material provided with a "reactive" metal is heated, a bonding interaction occurs under favourable conditions which may be based on partial reduction of the oxide present in the ceramic material with a simultaneous oxidation of the reactive metal.

ceramic materials

metal foils

In order that the invention may be clearly understood and readily carried into effect, reference will now be made, by way of example, to the accompanying drawing, in which:—

Figures 1 to 7 are diagrammatic sectional views illustrating respective examples of the bonding of a ceramic part to a metallic part.

Referring now to the drawings and, more particularly to Figure 1, there are illustrated a first metallic layer 3, a second metallic layer 1, and an intermediate metallic layer 2, which are arranged between a metal body M and a ceramic body K, said layers being either loosely superposed or firmly connected to one another. The layer 1 consists of a solder metal, preferably hard solder, by way of example, a gold-nickel sheet or foil preferably having a thickness of at least 25 microns. The layer 3 contains reactive metal and may be formed of a zirconium foil preferably having a thickness of at least 10 microns. Arranged between the layers 1 and 3 is an intermediate layer 2, for example, a nickel foil preferably having a thickness of at least 125 microns. Upon adequate heating, the gold-nickel layer 1 will fuse and solder the metal body M to the layer 2 forming a eutectic mix or alloy with part of the metal of the layer 2, that is in this case with the

solder

foil

125 foil

nickel. The zirconium layer 3 forms together with the nickel layer 2 to eutectic which will fuse below the temperature at which the heat treatment is performed, the eutectic reacting by bonding with the ceramic material K. The portion of Figure 1 below line A—A shows the alteration of the metal layers under the influence of heat. The layer 1^1 is thicker than the layer 1 and has absorbed a quantity of material from the layer 2 corresponding to the increase in thickness of layer 1. Analogously, the layer 3 has increased to layer 3^1 by absorbing metal from the intermediate layer 2. The thickness of the solid separating layer 2^1 after bonding has thus become less than that of layer 2 prior to bonding. The layer 2^1 remains solid both during and after the heat treatment.

The presence of the reactive metal in the layer 3^1 and the absence of this metal in layer 1^1, owing to the barrier action of the separating layer 2^1, is responsible for the difference in hardness and brittleness between the layers 1^1 and 3^1 respectively. The layer 1^1 is softer or more ductile than layer 3^1 and can therefore take up forces resulting from the different coefficients of expansion of the ceramic K and metal M when the respective temperature changes occur (changes due to cooling of the bond, variations in the operating conditions and so forth) without destroying the bond.

Figure 1 further shows that the thickness of the separating layer is a function of the type and thickness of the adjacent metal layers and also of the bonding temperatures employed. Too thin a separating layer might, under certain conditions and when operating at a given temperature, completely react with the two fused adjacent zones so as to form an alloy, all metal components then forming a homogeneous melt which would cause embrittlement and/or hardening of the complete bonding area, which is, of course, to be avoided. The thickness of the individual layers may be determined empirically by known metalography techniques.

The bonding temperature must always be lower than the melting point of the metal in the layer 2. The bonding temperatures must, however, be higher than this melting point of the lower-melting alloys located between the metal of the layer 2 and the solder on the one hand, and the metal of the layer 2 and the reactive metal on the other. In practice, an intermediate layer 2 formed of silver is suitable for bonding operations up to 900° C, of copper up to 1000° C and of nickel or cobalt up to 1400° C.

As intermediate layer

Figure 2 illustrates the principle of a bonding operation employing only two layers 9, 10 between the two bodies M and K. In this case, the reference numeral 9 designates the second metallic layer, *i.e.* the solder metal, which provides the solid separating layer 9^1 during the heating operation, and at the same time the alloy zone 9^{11} with the metal body M. The first metallic layer 10 is again formed by the reactive metal which will bondingly react with the ceramic body K. The explanation of the portion of Figure 2 located below the dashed centre-line B—B is entirely analogous to that of Figure 1. The layer 9^{11} is the alloy zone which forms between the metal M and the separating layer 9^1.

Figure 3 illustrates application of a soldering technique according to Figure 1 to a ceramic tube 13 which has both its ends closed by the metal covers 11 and 12. Arranged between the ceramic tube 13, which may alternatively be a solid rod, and the covers 11 and 12 are the metal foils 14, 15, 16 and 14^1, 15^1, 16^1. The foils 16, 16^1 containing the reactive metal bear directly against the tube 13 and have the two intermediate layers 15 and 15^1 and the layers of solder metal 14 and 14^1 superposed.

Figure 4 illustrates the manner in which two superimposed ceramic cylinders 19 and 21 may be connected by an intermediate metal ring or rod 20. The thickness of the intermediate metal layer 20 is not a limiting factor so that short tubes or discs may also be employed in the embodiment of Figure 4. This assembly is then closed at its opposite ends by the cover plates 17 and 18 formed of metal. The composites consisting of the metal layers 22, 23 and 24 correspond insofar as arrangement and composition is concerned to the layers 14, 15 and 16, respectively, of Figure 3 and are inserted between each metal and ceramic body. The bonding effect is similar to that heretofore described.

Figure 5 represents a tubular arrangement wherein a ring 26 formed of a ceramic material is located between two outer metallic rings 25 and 27. The metal layers 28, 29 and 30 are similarly arranged as the layers 14, 15 and 16 of Figure 3. Such an arrangement is particularly suited, by way of example, to the electric insulation of two metallic evacuated hollow bodies which have applied to them different electric potentials.

Figure 6 illustrates the manner in which a metal lead-in 32 provided with a bore 33 is connected to a ceramic plate 31 provided with a counterbore 33*a*. Analogous to the examples hereinabove disclosed, the metallic body 32 and the ceramic portion 31 have arranged between them the composite consisting of metal layers 34, 35, 36; reference numeral 34 designating the solder layer, reference numeral 35 the intermediate layer and reference numeral 36 the layer containing the reactive metal. Figure 7 substantially corresponds to that of Figure 6 and illustrates how a solid metal lead-in 38 may be bonded to a ceramic plate 37 using metal layers 39, 40, 41 similar to the layers 14, 15, and 16 of Figure 3.

different solder, No Ah

The embodiments represented in Figures 3 to 7 are all shown to be butt-joined because this type of bonding is particularly sensitive as regards expansion of the materials under the influence of heat, thereby best illustrating the advantages of the present method. Naturally, other types of joints may be produced in this manner.

Examples of the present method will now be given in greater detail.

EXAMPLE I

metal

ceramic

A metal body formed of a material ~~belonging~~ to the iron-nickel-cobalt group was employed, ~~the ceramic body~~ consisting of a commercial Al_2O_3 sintered ceramic material of a purity ~~of about~~ 97%. The metal body and the ceramic body were each provided with a plane surface having the same area and at which the bonding operation was to be performed. The arrangement of the metal layers or foils was effected according to Figure 1. The layer 1 in Figure 1 consisted of a hard solder foil of a thickness of 25 microns formed of 81.5% gold and 18.5% nickel (melting point ~~approximately 950° C.~~). The layer 3 consisted of commercially pure zirconium and ~~had a thickness of 10 microns~~. The intermediate layer 2 consisted of pure nickel (melting point about 1455° C.). The entire assembly was uniformly heated to approximately 1000° C. during the course of two hours in a vacuum (15^{-5} Torr.) and then kept at this temperature for ten minutes. This caused a nickel-zirconium eutectic (melting point approximately 961° C.) to be formed. As zirconium and nickel can react with each other only in those relative amounts which correspond to the bonding temperature, the thickness of the layers 2 and 3 required can be determined from the known two-component systems in this and analogous cases so that sufficient zirconium is present for the reaction with the ceramic material. Sufficient surplus nickel is required that only a portion thereof is used for the formation of the alloy while the balance will remain unfused and unalloyed as the separating layer 2^1. On the other side of the separating layer, the eutectic in layer 1^1 can remove only little nickel because the common melting point immediately increases due to its incorporation or alloying. After the bonding operation, it is found that part of the intermediate layer 2 consisting of nickel is alloyed with the zirconium in layer 3^1 and, in layer 1^1, with the gold-nickel. The non-fused balance, that is the separating layer 2^1 provides, by virtue of its melting point of 1455° C, an effective barrier between the brittle nickel-zirconium and the soft bonding and zirconium-free nickel-gold which latter retains its full ductility after the operation.

Au-Ni foil

metal film Ni intermediate layer

EXAMPLE II

The assembly of ceramic and metal bodies is carried out as in Example I. The metallic layers are formed of titanium for reactive layer 3, copper for boundary layer 2 and a silver-copper alloy containing 28.5% copper for solder layer 1. Titanium and copper will form a eutectic at about 850° C from which the fused titanium will react with the ceramic material. On the metal side, the eutectic of silver-copper will fuse at approximately 900° C. The melting point of the pure copper is about 1083° C. At a bonding temperature of about 950° C both eutectics are accordingly fused while separating layer 2 remains in the solid state so as to prevent penetration of titanium into the fused silver-copper of solder layer 1.

EXAMPLE III

An Al_2O_3 sintered ceramic material having a purity of at least 97% is bonded to a metal body formed of gold (pure gold). For this purpose, a Ti foil (commercially ~~pure~~) having a thickness of 10 microns and a Cu foil of a thickness of 100 microns are arranged between the ceramic body and the metal body (according to Figure 2). The Ti foil corresponds to the layer 10 in Figure 2, and the Cu foil to the layer 9. The entire arrangement was heated to approximately 950° C in gettered welding argon within two hours and then kept at this temperature for ten minutes. After cooling, the composition of the layers corresponds to the layer arrangement shown below the dashed centre-line B—B of Figure 2. The gold metal body has its surfaces alloyed with the copper. On the side facing the ceramic material, a copper-titanium alloy has formed, which is bonded to the ceramic material and the copper separating layer.

Au substrate
Ti foil
Cu foil
Al_2O_3 ceram

The composite suitable for the performance of the method is a multi-layer assembly comprising at least two metals, the assembly consisting of a metal reacting with a ceramic material, such as zirconium or titanium, on the one hand, and of a hard solder, such as gold, gold-silver, silver-copper, or gold-nickel, ~~on the other~~. This multi-layer assembly may consist of loosely superposed or firmly bonded layers connected by plating, vaporising of galvanising one metal layer on another.

Au+Au-Ag solder

It may in many cases be of advantage not to use the reactive metal as a foil, but to apply it to the adjacent layer or the ceramic material by either mechanical, electrolytical or vacuum-type methods as previously stated.

The bodies formed by the present method, particularly those in the form of sintered ceramic material consisting of high-percentage Al_2O_3 or BeO and metal bodies bonded thereto, particularly formed of Fe—Co—Ni alloys of various types, are particularly suited for high-frequency application. High frequency equipment provided with parts manufactured by this method is particularly distinguished by its thermal resistance. The pos-

sibility of constructing such equipment while no longer being restricted to employing ceramic and metal bodies having similar coefficients of expansion within the working temperature range constitutes a principal advantageous use of the present method.

WHAT WE CLAIM IS:—

only 2 interior layers

1. A method of bonding a ceramic part to a metallic part, comprising disposing between said ceramic part and said metallic part first and second different metallic layers such that the first layer, ~~which comprises a first~~ metallic material capable of reacting with, and bonding itself to said ceramic part, is disposed adjacent said ceramic part and that the second layer, which comprises a second metallic material capable of bonding itself to said metallic part, is disposed adjacent to said metallic part, heating the ceramic part, the metallic part and the layers in an inert atmosphere or in vacuo so as to bond the ceramic part to the metallic part, and causing there to be present during the heating operation a solid metallic separating layer separating molten, first metallic material adjacent to said ceramic part from molten, second metallic material adjacent to said metallic part, eutectics being formed between said separating layer and said molten, first metallic material and between said separating layer and said molten, second metallic material or said metallic part, the heating being performed at a temperature lower than the melting point of said separation layer and higher than the melting point of each of said eutectics.

2. A method as claimed in claim 1, wherein said second metallic material is a metal capable of forming a eutectic with said metallic part, said separating layer being formed by part of said metallic layer.

3. A method as claimed in claim 1, and including locating between said first and second different metallic layers before the heating operation a third metallic layer having its melting point above said temperature and capable of forming eutectics with said first metallic material and with said second metallic material, said separating layer being formed by said third metallic layer, and said second metallic material being a hard solder.

4. A multi-layer assembly for use in a method as claimed in any one of claims 1 to 3, comprising, at one face of said assembly, said first metallic layer which comprises a first metallic material capable of reacting with, and bonding itself to, said ceramic part and, at the opposite face of said assembly, said second metallic layer which comprises a second metallic material capable of bonding itself to said metallic part.

5. A multi-layer assembly as claimed in Claim 4, wherein the layers are firmly bonded together.

6. A multi-layer assembly as claimed in claim 4 or 5 and for use in a method as claimed in Claim 3, wherein said third metallic layer, which has its melting point above said temperatures and is capable of forming eutectics with said first metallic material and for high frequency application. High frewith said second metallic material, is intermetallic layers.

7. A method of bonding a ceramic part to a metallic part, substantially as hereinbefore described with reference to any Figure of the accompanying drawings, and/or any one of examples I to III.

8. A multi-layer assembly, substantially as hereinbefore described with reference to any Figure of the accompanying drawings, and/or any one of Examples I to III.

9. A ceramic part and a metallic part bonded together by a method as claimed in claim 1, 2, 3, or 7.

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979811 COMPLETE SPECIFICATION

1 SHEET

This drawing is a reproduction of the Original on a reduced scale

soldered Au-Ni

optional layer Ni

reactive metal Ti or Zr foil

Fig. 1

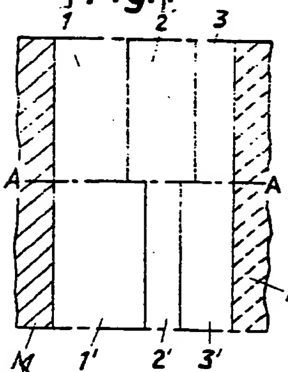


Fig. 2

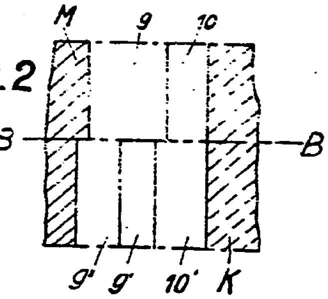


Fig. 3

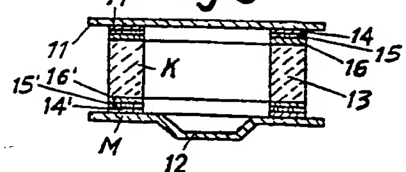


Fig. 4

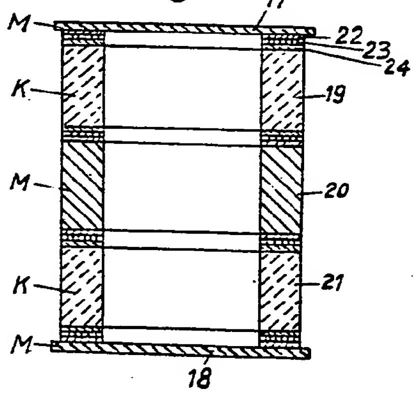


Fig. 5

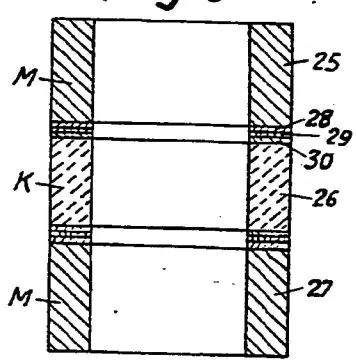


Fig. 6

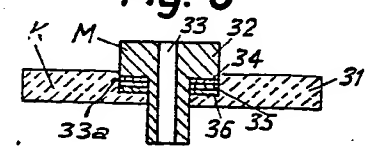
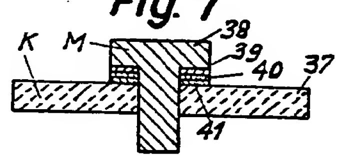


Fig. 7



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